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LA-UR--89-48

DE89 006546

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Received by OSTI

FEB 0 6 1989

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AUTHOR(S) Leon J. Radziemski, Noshir M. Khambatta, John A. Oertel, and Robert Silk - New Mexico State University, Dept. of Physics, Las Cruces, NM 88003

Joseph M. Mack - Los Alamos National Laboratory, Los Alamos, New Mexico 87544

Dushan Mitrovich - #3 Mulberry Loop, Cedar Crest, NM 87008

SUBMITTED TO O/E Lase 1989 - Los Angeles, California, January 15-20, 1989

MASTER

Los Alamos

Los Alamos National Laboratory
Los Alamos, New Mexico 87545

On the use of multiple photon processes in krypton for laser guiding of electron beams

by

Leon J. Radziemski, Noshir M. Khambatta,
John A. Oertel, Robert Silk

New Mexico State University
Department of Physics
Las Cruces, NM 88003

and

J. M. Mack
GRP P-4, MS E554
Los Alamos National Laboratory
Los Alamos, New Mexico 87545

and

Dushan Mitrovich*
#3 Mulberry Loop
Cedar Crest, NM 87008

ABSTRACT

Neutral krypton atoms were excited from the ground state $4p^6\ {}^1S_0$ to the $4p^5\ 6p[3/2]_2$ state by a resonant two-photon absorption from a line-narrowed ArF excimer laser operating at 193.41 nm. A third photon, absorbed while the atom is in the excited state, ionizes it. Excited state and ion densities were theoretically computed using a standard rate-equation analysis. The irradiance levels used ($1\text{-}5\times10^8$ W/cm^2) were too low for significant ground and excited state ac Stark and Rabi effects. The photon detection system was calibrated with a standard tungsten lamp. Ion signals were measured with known electrical components. The resonance results were compared with predictions of non-resonant ionization based on a standard formulation. The ion and excited state densities have been used with a modified electron beam propagation code (IPROP) to model such propagation in a low pressure laser-excited krypton channel. The modifications included the effects to field ionization of the excited krypton atoms. Implications for guiding of e-beams using ArF excited krypton are discussed.

*Some of this work was performed while DM was at Mission Research Corp., Albuquerque, NM 87106.

1. INTRODUCTION

Since the availability of high-powered lasers, multiphoton spectroscopy has emerged as a powerful tool for studying the properties of excited and Rydberg states of atoms and molecules.[1,2] We were motivated to investigate multiphoton excitation and ionization in atomic krypton, for use as a "guiding gas" to channel an 0.5-10 kA electron beam. Currently such guiding is performed by photoionizing low pressure benzene with two photons from a KrF laser.[3,4] Krypton would be a more benign and easily modeled working gas. It is excited resonantly using a wavelength narrowed ArF laser operating at 193.41 nm.[5] Some of the results described below are also discussed in References 6, and 7.

2. THEORY

The fundamental quantity, the two-photon absorption cross section $\sigma_o^{(2)}$ is given by[5,6,7]

$$\sigma_0^{(2)} = (2\pi)^3 (\alpha)^2 (h\upsilon)^2 |M|^2 \tag{1}$$

$$M = \sum_i \frac{\langle f | r^{\lambda} | i \rangle \langle i | r^{\lambda} | g \rangle}{(E_g - E_i + h\upsilon)} \tag{2}$$

We rewrite the matrix element M in terms of the familiar dipole transition matrix element (μ) as

$$|M|^2 = \left| \frac{2 \mu_1 \mu_2}{e^2 |E_k - h\upsilon|} \right|^2 \tag{3}$$

where a "single-path approximation" has been utilized. E_i is the energy of the single intermediate state. The dipole transition matrix element is by definition

$$\mu = \sqrt{e^2 |\langle \alpha LSJM_J | \hat{R}^{(el)} | \alpha' L' SJ' M'_J \rangle|^2} \tag{4}$$

where

$$|\langle \alpha LSJM_J | \hat{R}_1^{(el)} | \alpha' L' SJ' M'_J \rangle|^2 = (2J'+1) \sum_q \begin{pmatrix} J & 1 & J' \\ -M_J & q & M'_J \end{pmatrix}^2 \frac{A^{(1)}}{\alpha^2 (0.667)} \frac{3}{\Delta E} \tag{5}$$

where $|\langle|R^{(el)}|\rangle|^2$ is in units of a_o^2, (a_o being the Bohr radius), $(\quad)^2$ are the 3j-symbols of interest. $A^{(\ell)}$ is the transition probability (in s^{-1}) between the levels (J', M'_J) and (J, M_J). σ is the transition wavenumber between these levels in cm^{-1}, and ΔE is the energy difference between these levels in units of Rydberg.[8]

Using these, the two-photon absorption cross-section $\sigma_o^{(2)}$ for the $4p^5 6p[3/2]_2$ level at a wavelength of 193.41 nm is evaluated to be $\sigma_o^{(2)} = 2.18\times10^{-37}$ cm^4. This corresponds to the two-photon coupling parameter $\alpha = 1.43\times10^{-32}$ cm^4/W. For comparison, two other published values of α are 2.34×10^{-31} cm^4/W (Ref. 5) and 2×10^{-32} cm^4/W (Ref. 9) for nearly comparable bandwidth ArF lasers.

The other fundamental quantity, the photoionization cross section σ_{pi} is computed for the $4p^5$ $6p[3/2]_2$ level of krypton by using the expression, based on a hydrogenic approximation,[7]

$$\sigma_{pi} = \frac{8\times10^{-18}}{Z\sqrt{U_I/R}\,(h\nu/U_I)^3} \tag{6}$$

yielding a numerical value of $\sigma_{pi} = 1.7\times10^{-19}$ cm^2. For comparison, the value from Ref. 5 is 3.2×10^{-19} cm^2.

For comparison with the nonresonant ionization, we note that as long as perturbation theory is applicable, the k-photon ionization can be described in terms of the generalized cross-section, σ_k (in units of cm^{2k} s^{k-1}) and the photon flux F ($cm^{-2}s^{-1}$), by defining the transition probability per unit time W_k as

$$W_k = \sigma_k F^k \qquad (s^{-1}) \tag{7}$$

The probability of ionization P_k is then given by the integral

$$P_k = \int_o^\infty \sigma_k F^k\, dt \tag{8}$$

while the number of ions produced at the end of the pulse (presuming single electron ejection) is

$$N_I = N_o\,(1 - \exp(-P_k)) \tag{9}$$

We consider a temporal pulse shape that is Gaussian in time, defined by[10]

$$I(t) = I_0 \exp(-t^2/\tau_0^2) \quad (10)$$

The pulse width at half maximum is defined to be

$$\tau_p = 2 \tau_0 (\ln 2)^{1/2} \quad (11)$$

and the average pulse irradiance is related to pulse fluence ϕ_p by

$$\bar{I} = \phi_p / \tau_p = 0.5 \, I_0 (\pi/\ln 2)^{1/2} \quad (12)$$

If we are far from the saturation region, Eq. (9) can be simplified to yield

$$N_i = N_0 \sigma_k (\bar{F_0})^k \tau_L \, g(k) \quad (13)$$

where F_0 is the averaged peak flux, τ_L is the duration of the laser pulse and g(k) is the pulse shape factor given by

$$g(k) = (\pi/4k \ln 2)^{1/2} [2(\ln 2/\pi)^{1/2}]^k \quad (14)$$

3. COMPUTATIONAL RESULTS

Using the resonant expressions one can solve the appropriate rate equations and compute the excited and ion number densities as shown in Table I. These are for two-photon resonant, three-photon ionization of atomic krypton.

Consider now the details of non-resonant 3-photon ionization of atomic krypton at 193.41 nm, at a pressure of 0.01 Torr and a temperature of 297°K.

At $\,$ = 10 mT; T = 297°K $\qquad$ $N_0 = 3.252\times10^{14}$ atoms/cm^3

At I = 10^8 W/cm^2 $\qquad$ $F_0 = 9.713\times10^{25}$ photons/cm^2-sec

$\tau_L = 12.2$ ns $\qquad g(3) = 0.51$

We use $\sigma_3 = (1 \times 10^{-82})$ cm^6-s^2 (Based on ref. 11). This is within a few % of the value quoted by McGuire[12] for circular polarization. This yields for N_i from eq. (13)

$$N_i = (3.252 \times 10^{14})(1 \times 10^{-82})(9.713 \times 10^{25})^3(12.2 \times 10^{-9})(0.51)$$

$$= 1.85 \times 10^2 \ cm^{-3} .$$

However for linearly polarized light, the generalized cross sections $_3$ are two orders of magnitude higher than those for circularly polarized light so N_i may now range from 1.85×10^2 to 1.85×10^4 atom/cm^3 for the same initial conditions. Recall that at 10^8 W/cm^2, under all similar conditions as above, for a two-photon resonant three-photon ionization of atomic krypton (see Table I) $N_i = 2.247 \times 10^8$ cm^{-3}.

Table I: Computed ground state (N_1), excited state (N_2) and ion number (N_i) densities for p(Kr) = 10 m Torr and various irradiance values.

I (W/cm^2)	N_1 (cm^{-3})	N_2 (cm^{-3})	N_i (cm^{-3})
1.0×10^6	3.251×10^{14}	2.334×10^5	2.396×10^2
1.0×10^7	3.251×10^{14}	2.313×10^7	2.382×10^5
1.0×10^8	3.251×10^{14}	2.120×10^9	2.247×10^8
1.0×10^9	3.249×10^{14}	1.021×10^{11}	1.369×10^{11}

4. EXPERIMENTAL RESULTS

The experimental results are discussed in detail in Refs. 6 and 7. Here we summarize possible effects upon the cross sections. The computed and measured values of N_2 agree well on average (except for the two lowest pressures) when a value of $\sigma_0^{(2)} = 5.4 \times 10^{-37}$ cm^4 (corresponding to a two-photon coupling parameter $\alpha = 3.5 \times 10^{-32}$ cm^4/W) is used. We find that on average, an adjusted $\sigma_{pi} = 4.39 \times 10^{-19}$ cm^2, would bring theoretical N_i results into agreement with the experimental ones. The uncertainties in the experimentally determined cross sections are ±50%.

5. LASER REQUIREMENTS

We here calculate the energy needed to provide a required line density of ions for electron beam guiding. Consider a 500 A, 5 ns pulse. The number of electrons is 1.56×10^{13}. The beam volume is, assuming a length of c x 5 ns = 150 cm, and a radius of 3mm, 423 cm^3. Hence the electron density is 3×10^{9} cm^{-3}.

The required laser-produced ion density for a charge neutralization fraction f = 0.1 is 3.7×10^{10} cm^{-3}, which corresponds to a line density of 1×10^{10} cm^{-1}. The beam line density is ten times as great.

Using ArF at 193 nm krypton p = 10 mT, an irradiance of 2.5×10^{8} W cm^{-2} gives the required line density (Table I). The corresponding pulse energy is

$$E = I\,A\,t = 2.5\times10^{8} \times \pi(.3)^2 \times 12 \times 10^{-9}$$

$$= 8.46\ \mathrm{J}.$$

For a 10 kA beam, the energy required is 23.7 J. If field ionization from excited 6p states works, the irradiance requirements are reduced to 5.3×10^{7} and 1.48×10^{8} W cm^{-2}, and the corresponding energies to 1.8 or 5.0 J.

6. BEAM GUIDING CALCULATIONS

An electron beam propagation code (IPROP) was modified to include Rydberg excitation and collisional ionization physics. It is a multispecies, relativistic, particle-in-cell simulation code with a full electromagnetic field solver in 3 dimensions, with the azimuthal coordinate analyzed into Fourier modes. A given length of beam can be followed for long distances by using a reference frame moving with the beam. Beam electrons, plasma electrons, and plasma ions comprise three separate species. Ground state krypton atoms can be ionized collisionally by beam electrons, and Rydberg atoms collisionally by all electrons (depending on cross section), and also by electric field ionization. All Rydberg atoms are assumed to have the same principal quantum number. An ionization event is treated as a simultaneous addition of a macro-electron and a macro-ion to their respective species, at the location of the event. A corresponding decrease in the source atom density is made. Simulations compute the composite ionization process, the electrodynamics of plasma electrons being expelled from the channel, and the guiding efficiency of the electron beam within the channel.

The essential results are as follows:

1. The field-ion production rate is a very sensitive function of radial field and quantum state.

2. Control of that production rate will be very difficult, although unusual spatial ion distributions could be generated (i.e. solenoidal).

3. A ten meter run with no background ions, has been finished. The beam propagated 10 meters, but the charge transport is not known at this time.

4. A ten meter run, with three-photon produced ions, is now being made.

7. CONCLUSIONS

Electron beam guiding over distances of tens of meters in low pressure krypton is feasible with sufficiently high ArF energy. This energy is significantly above that now available from standard laboratory excimers.

8. ACKNOWLEDGEMENTS

We acknowledge support under the Department of Energy contract DE-RA03-86SF16496 and the Department of Defense instrumentation grants DAG-29-85-G-0012 and DAAL03-86-G-0119. The able experimental assistance of Dr. Robert Liefeld (NMSU), Dr. Abhijit Biswas (NMSU), and Todd Kaupila (LANL) is appreciated.

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